sion. I succeeded in 1895 in obtaining some evidence as to the separa-
tion of gold from its solution in metallic lead by electrolysis through a
glass septum.* This is, however, only indirectly connected with the
electrolysis of alloys.]

"On Certain Properties of the Alloys of the Gold-Copper Series." By Professor Sir W. Roberts-Austen, K.C.B., F.R.S., and T. Kirke Rose, D.Sc. Received and read May 10, 1900.

[Plate 1.]

Notwithstanding the extraordinary importance from a technical
point of view of the members of this series, which constitute the gold
coinages of the world, singularly little is known respecting either their
molecular constitution or even their physical constants. Both the
authors of this paper possess unusual facilities for studying them, and
they felt that time should not be lost in beginning a systematic exami-
nation of the series. The other alloys used for coinage have, on the
other hand, not been so neglected. Many years ago one of us,† in
submitting his first paper to this Society, gave a curve representing
the freezing points of the members of the silver-copper series. This
curve, corrected in accordance with more recent work and interpreted
in a modern way, proved to be one with two branches meeting at a
point where the eutectic alloy of the two metals occurs. The presence of
the eutectic has also been since readily detected in standard silver and
in several other members of the series, and possesses a melting point of
778°. As is well known, different portions of a mass of any of the
solidified alloys of the silver-copper series, except the eutectic alloy,
exhibit divergences in composition which usually amount to about two
or three parts in a thousand.

The gold-copper series, on the other hand, has long enjoyed a
reputation for homogeneity, and it was supposed that the variations in
the composition either of the alloy which contains 916·66 parts of gold
in 1000, and is used for the coinage of the Empire, or of the alloy
which contains 900 parts of gold in 1000, and is one adopted by the
Latin Union and in the United States of America, need not exhibit
greater divergences than 0·1 part in 1000. It was, moreover, believed
that such a divergence was not the result of any systematic molecular
grouping. This view was shaken by one of us‡ in 1895, when
evidence was obtained by chemical analysis that in the case of a gold-

* Third Report to the Alloys Research Committee, ' Proc. Inst. Mech. Engineers,
1895, p. 240.
† Roberts-Austen, ' Roy. Soc. Proc.,' vol. 23 (1874), p. 481.
‡ Rose, ' Chem. Soc. Journ.,' vol. 67, 1895, p. 552.

copper alloy containing 0·2 per cent. of impurity a certain amount of the gold was driven to the inside of the mass by solidification. Corroborative evidence was subsequently obtained by the aid of the cooling curves afforded by the recording pyrometer, a description of which has already been submitted to this Society.

To decide the point finally it was desirable to show to what group of alloys the gold-copper series belongs, and in particular to determine whether the freezing points of the various alloys would lie on a single continuous curve connecting the freezing point of gold with that of copper.

Freezing-point curves were accordingly taken by the recording pyrometer of a comprehensive series of alloys. In each case 100 grammes of the alloy were employed, and the thermo-couple, protected by a very thin clay tube, was inserted in the molten mass, which had been previously thoroughly stirred. The rate of cooling was prolonged as much as possible by allowing the crucible and its contents to remain in position in the gas furnace in which the melting had been effected. The freezing points of this series have, so far as we are aware, never been published, except a few at the copper end by Heycock and Neville.* MM. Charpy and Riche have, however, recently stated that the curve of fusibility of the alloys of gold and copper consists of two branches meeting at a point corresponding to the eutectic alloy which, according to these experimenters, contains 55 per cent. of gold, alloyed with 45 per cent. of copper, and fuses at 940°.† This conclusion is not confirmed by the results of our experiments, which are given in the accompanying table and are plotted in the curve, fig. 1.

FIG. 1.
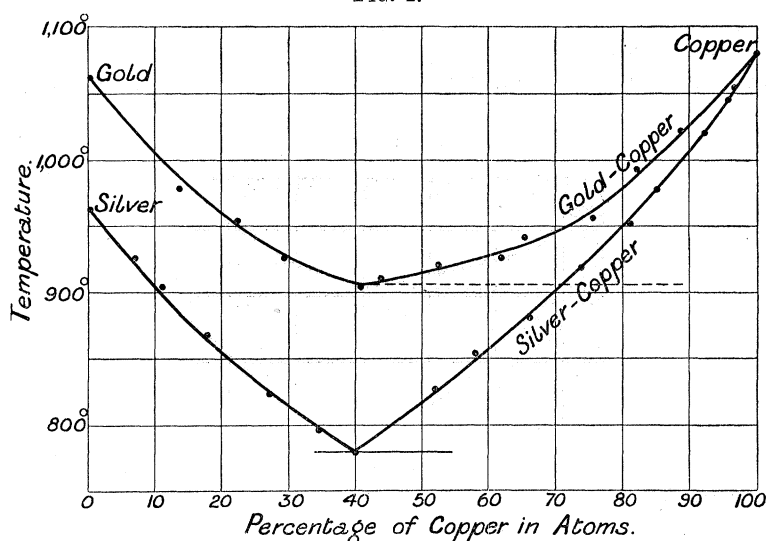
Freezing points of Alloys of Gold and Copper.

Percentage weight of gold.	Number of atoms of gold per 100 of alloy.	Freezing point.
100 ·00	100 ·00	1063° C.
95 ·00	86 ·00	979
91 ·60	77 ·80	951
90 ·07	74 ·45	946
88 ·06	70 ·32	926
82 ·05	59 ·49	905
79 ·97	56 ·19	907
73 ·83	47 ·54	916
66 ·26	38 ·69	928
62 ·20	34 ·58	941
52 ·03	25 ·84	957
51 ·87	25 ·72	963
40 ·45	17 ·88	994
27 ·70	10 ·96	1022
11 ·15	3 ·87	1059
0	0	1083

The initial freezing points of the gold-copper alloys are easy to determine, but the subsidiary or eutectic points are very difficult to detect even if a sensitive autographic recorder is employed. We have great confidence in our conclusion that the alloy containing about 82 per cent. of gold and 18 of copper, and not the one which contains 55 per

cent. of gold, is really the eutectic. The reason, apart from micro-graphic evidence, is not only that the freezing point of the 82 per cent. gold alloy is lower than that of any other member of the series, but the autographic record reproduced in fig. 2 shows that the angles at A

Fig. 2.
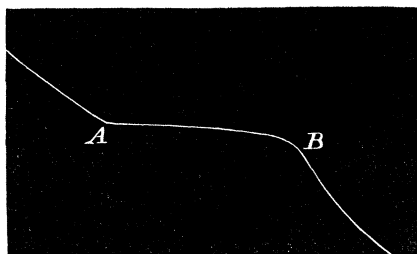
and B, where the solidification begins and ends, are quite sharp, while the portion between A and B, which represents the actual solidification of the alloy, is horizontal. Neither of these conditions are met with in the autographic records of the other alloys of the series. Moreover, the fracture of the 82 per cent. alloy is conchoidal, as in the case of a great number of other eutectics, owing to the extremely fine state of division of the constituents, which makes these alloys appear to be homo-geneous. The exact composition of the eutectic is, however, difficult to determine.

A comparison of the freezing-point curve of the gold-copper with that of silver-copper alloys shows that there are striking similarities when the number of atoms in the alloys are taken as abscissæ. It was shown by Levol[*] as long ago as 1852 that the only homogeneous alloy of silver and copper corresponded in composition with the formula Ag_3Cu_2, and Heycock and Neville[†] confirmed the anticipation of one of us,[‡] which was not verified at the time, that it would prove to be the eutectic of the series. In the gold-copper series the alloy contain-ing 59·49 atoms of gold and 40·41 atoms of copper has a lower freezing point than any other alloy examined, although it is hardly to be distinguished from the alloys containing a little more copper. The curve of fusibility of the series is much more rounded near this point than that of most binary alloys, and bears a superficial resemblance to that of two substances forming a continuous series of mixed crystals, but micrographic study of the series conclusively shows that it possesses a eutectic.

* Levol, 'Annales de Chim. et de Phys.,' vol. 36 (1852), p. 193 ; vol. 39 (1853), p. 163.

† 'Phil. Trans.,' A, vol. 189 (1897), p. 25.

‡ Roberts-Austen, loc. cit.

It has been shown by Osmond,* moreover, that silver and copper are each capable of holding a small percentage of the other in solid solution, but that if both metals are present in considerable amounts, the two solidified solutions exist side by side. It is evident therefore that they form an interrupted series of "mixed crystals," and that the substance first solidified in cooling a solution of one metal in the other is not a pure metal, but an isomorphous mixture of the two metals containing only a small percentage of one of them. This conclusion agrees well with the general shape of the curve of fusibility of the silver-copper series, and the still greater concavity of the curve of fusibility of the gold-copper series suggested that a similar condition of things is here met with, but that the gap in the series of mixed crystals is much smaller, and that the mutual solubility of these two metals is greater.

Microscopic examination of the alloys of gold and copper affords evidence that this is really the case, but appears to point to the conclusion that more copper can be dissolved in gold than gold in copper. Alloys containing only a small percentage of copper consist of large crystals similar in shape to those seen in pure gold, and showing no signs of cement between them. They differ from those of pure gold in their colour, which is reddish or reddish-brown, after treatment with nitrohydrochloric acid. When magnified 1580 diameters these crystals show a minutely granular structure which resembles that of pure gold, and affords no evidence of separation into two constituents. Even in standard gold containing only 91·6 per cent. of gold the structure is nearly the same, and is not unlike that of the ground mass of standard silver containing 92·5 per cent. and 7·5 per cent. of copper prepared in a similar way. On the other hand, the alloys containing less gold than the eutectic show crystals of copper set in a matrix which consists apparently of the eutectic.

The following examples of photomicrographs of the series are shown in Plate 1 :—

Fig. 1, Plate 1, represents the characteristic surface of a small ingot of standard gold. The structure was not developed by etching, and the magnification is only 4·5 diameters.

Fig. 2 is a polished section of standard gold etched by immersion for about 15 seconds in a boiling mixture of equal parts of nitric and hydrochloric acid. The magnification is, as in the case of No. 1, 4·5 diameters, and the structure consists of sections cut in various directions by a plane passing through the crystals, of which the mass is composed. Fig. 3 is the eutectic of the gold-copper series ; it contains 80 per cent. of gold and 20 per cent. of copper etched as in the case of the alloy shown in fig. 2 ; the magnification is, however, 1580 diame-

* 'Bull. de la Soc. d'Encouragement,' 5th Series, vol. 2 (1897), p. 837.

ters, which reveals the banded structure characteristic of a eutectic alloy.

Fig. 4 is standard gold etched as before and magnified 1580 diameters.

Fig. 5 is a section, etched as before, of an alloy containing 27 per cent. of gold, and 73 per cent. of copper. In it the presence of two distinct constituents can be seen. The darker portion, which has been readily attacked by the acid, is copper, and the lighter is mainly the eutectic. This fact is proved by fig. 6, which is a very high magnification (6300 diameters) of the lighter portion of fig. 5, and this on close examination reveals the presence of the laminated or banded eutectic.

Another resemblance between the series of gold-copper and silver-copper alloys is to be found in their relative tensile strengths. In both cases the eutectic alloys are extremely brittle, and have a lower tenacity than the other members of the series. In the case of gold, one of us* has shown that the tenacity of an unworked cast bar of pure gold 7·5 mm. wide and 5·2 mm. thick is 7 tons per square inch, the metal elongating 30·8 per cent. before rupture. Both tenacity and extensibility are greatly increased by the first additions of copper, the tenacity rising in the case of standard gold which contains 8·3 per cent. of copper to more than twice that of pure gold. Under similar conditions, however, we have found that the eutectic alloy of gold and copper has a tensile strength of only 7·87 tons per square inch, with an elongation of only 3·3 per cent. It is in fact about as brittle as pure gold alloyed with 0·24 per cent. of lead, which has an elongation of 4·9 per cent. We also determined the extensibility of the eutectic alloy of silver and copper to be only 2·2 per cent., and its tensile strength 29·1 tons per square inch. These are the first cases observed in which eutectic alloys appear to show less tenacity and extensibility than the other members of the series to which they belong. The eutectics of lead and tin, of copper and tin, and of iron and carbon are in each case the strongest alloys of the series, and are not at all brittle. The eutectic of the copper-zinc series is more extensible than any other member of its series, while its tenacity is considerable. The gold-copper and silver-copper alloys differ therefore from other alloys, which appear to be brittle and of low tensile strength only if they have passed through a pasty stage in solidifying, and possess two freezing points, the lower of which is that of the eutectic.

It is clear, from the results given above, that gold and copper cannot be expected to form a series of alloys of uniform composition, but will show evidence of liquation similar to that exhibited by silver and copper, though in a less degree. Much evidence on this point was obtained in the course of the preparation of the standard gold trial

* Roberts-Austen, 'Phil. Trans.,' A, vol. 179 (1888), p. 339.

plate, which contains 91·6 per cent. of gold and 8·3 per cent. of copper. This alloy was cast into flat bars of various dimensions, and assays were made on pieces cut from all parts of the plates into which the bars were rolled, care being taken to adopt all the precautions described by one of us with a view to ensuring accuracy in the determinations.* This enabled the limit of error to be reduced to 0·02 per 1000 on a mean of three assays. In all, nine plates were prepared before one of the necessary accuracy was obtained, and over 900 determinations of the proportion of gold present in the assay pieces were made. It was found that in general there was a tendency for the outside of the ingots to be richer in gold than the interior, but that this distribution was hardly so regular, and was not so pronounced as that observed in a contrary sense, in standard silver, in which case silver accumulates in the centre of the mass.

It may be added that the differences in composition of different parts of the gold bars, though small, are many times larger than the possible errors of assay.

The plates were of various dimensions, and were prepared from pure gold and electrodeposited copper, well stirred to ensure uniformity while in the molten condition, cast in iron moulds coated with carbon, and rolled out to a width of about 17·5 cm., a thickness of 1 mm., and a length of from 1 to 1·25 metres, the weight being from 3·7 to 4·6 kilos. Series of discs were then cut out in parallel lines, one down the centre of the plate, and two others distant 1 cm. from the edges. In the case of plate No. 1, intermediate series of discs were cut half way between the centre line and the edges. The means of all assays of each series taken from three typical plates were as follows, each result giving the mean of from 21 to 27 assays :—

	No. 1.	No. 2.	No. 3.
Left side	916·59	916·65	917·03
Left intermediate	0·65		
Centre line.	0·49	916·55	916·71
Right intermediate	0·66		
Right side	0·61	916·58	916·95
Mean of all assays	916·589	916·598	916·895
Greatest differences from mean (per 1000 parts)	{ +0·35 { −0·36	+0·14 −0·13	+0·60 −0·40
Difference between richest and poorest parts of the plate (per 1000 parts)..........	0·71	0·27	1·00

In the case of standard silver plates of the same size prepared in a similar way, the difference between the amounts of silver in the richest

* Rose, ' Chem. Soc. Journ.,' vol. 63 (1893), p. 704.

and poorest parts of the plate is usually from 1·0 to 3·0 parts per thousand, or three or four times as great as that in the case of standard gold. The poorest part in the gold plate is, however, always in the centre and the richest part at the outside.

The assays made on the " get " of the gold plates, the place on the top of the casting where shrinkage of the mass on solidifying is marked externally by a depression, showed that this part was usually richer in gold than any other part of the plate. These assays are not included in the means given above.

Conclusion.

It will be evident from the results given above, that when a small proportion of copper is added to gold, the alloy sets as a whole, and forms a solid solution. If small amounts of copper are successively added, the limit of solubility of that metal in gold is at length reached, and a eutectic separates, which forms the whole mass when about 82 per cent. of gold and 18 per cent. of copper are present.

Comparatively small additions of gold to copper saturate the latter, and the eutectic makes its appearance before the proportion of gold reaches 27 per cent. The composition of the eutectic corresponds approximately to 60 atoms of gold and 40 of copper, while the silver-copper eutectic also contains nearly 60 atoms of silver and 40 of copper. In other respects also, in the brittleness of the eutectic, in the limited mutual solubility of the two metals, and in the liquation which attends solidification, the gold-copper and silver-copper series resemble each other closely. The main difference is that copper appears to be more soluble in gold than in silver, so that the characteristics of the gold-copper alloys are less marked, and consequently have been less easy to detect.

" The Crystalline Structure of Metals." Second Paper. By J. A. EWING, F.R.S., Professor of Mechanism and Applied Mechanics in the University of Cambridge, and WALTER ROSENHAIN, B.A., St. John's College, Cambridge, 1851 Exhibition Research Scholar, Melbourne University. Received May 17,—Read May 31, 1900.

(Abstract.)

The investigations described in this paper deal principally with the phenomena of annealing. The first section of the paper describes experiments made in the hope of observing under the microscope the process of recrystallisation in strained iron. It is well known that
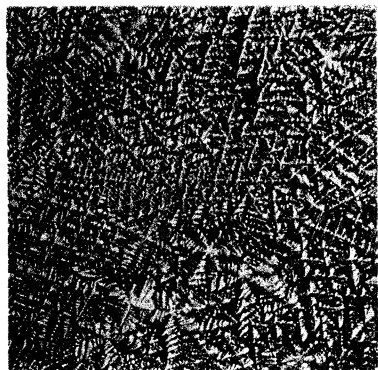
Fig. 1.   Surface of an Ingot of Standard
Gold, (unetched) x 4.5D.

Fig. 2.   Standard Gold. x 4.5D

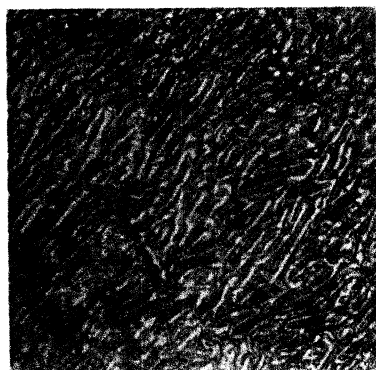Fig. 3. Eutectic, (80 per cent Gold,
20 per cent Copper) x 1580D.

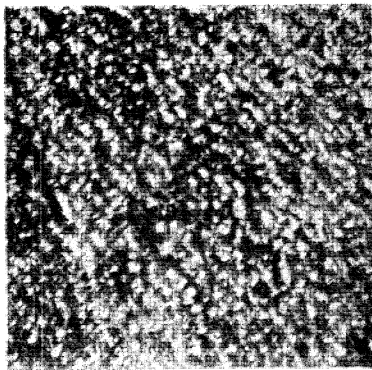Fig. 4.   Standard Gold. x 1580D.
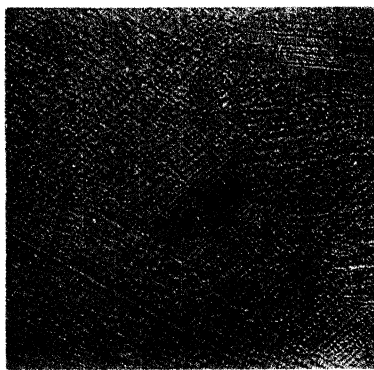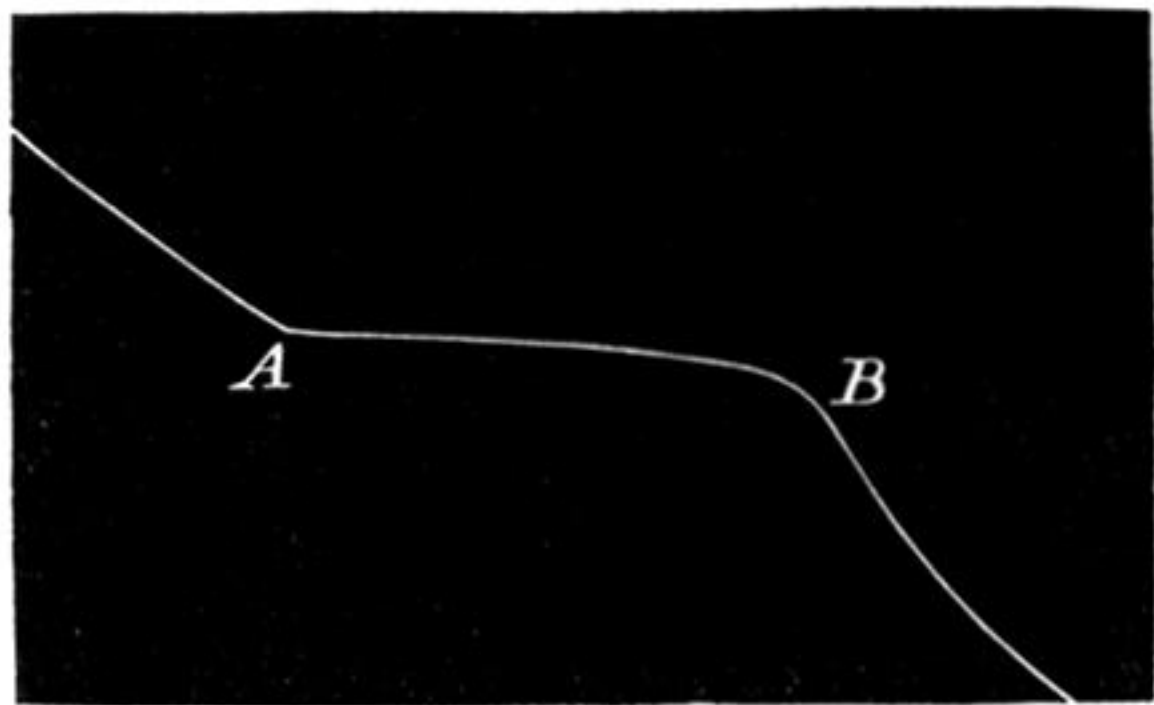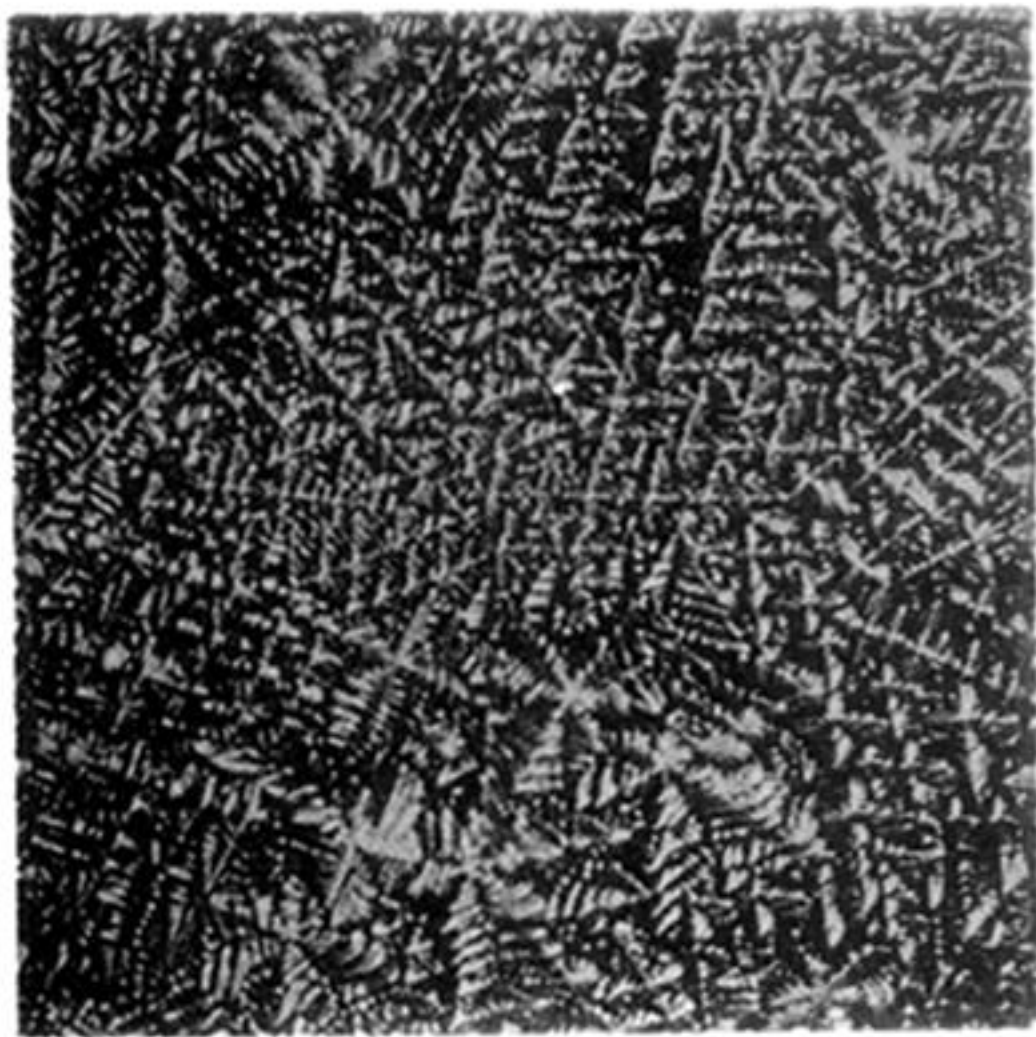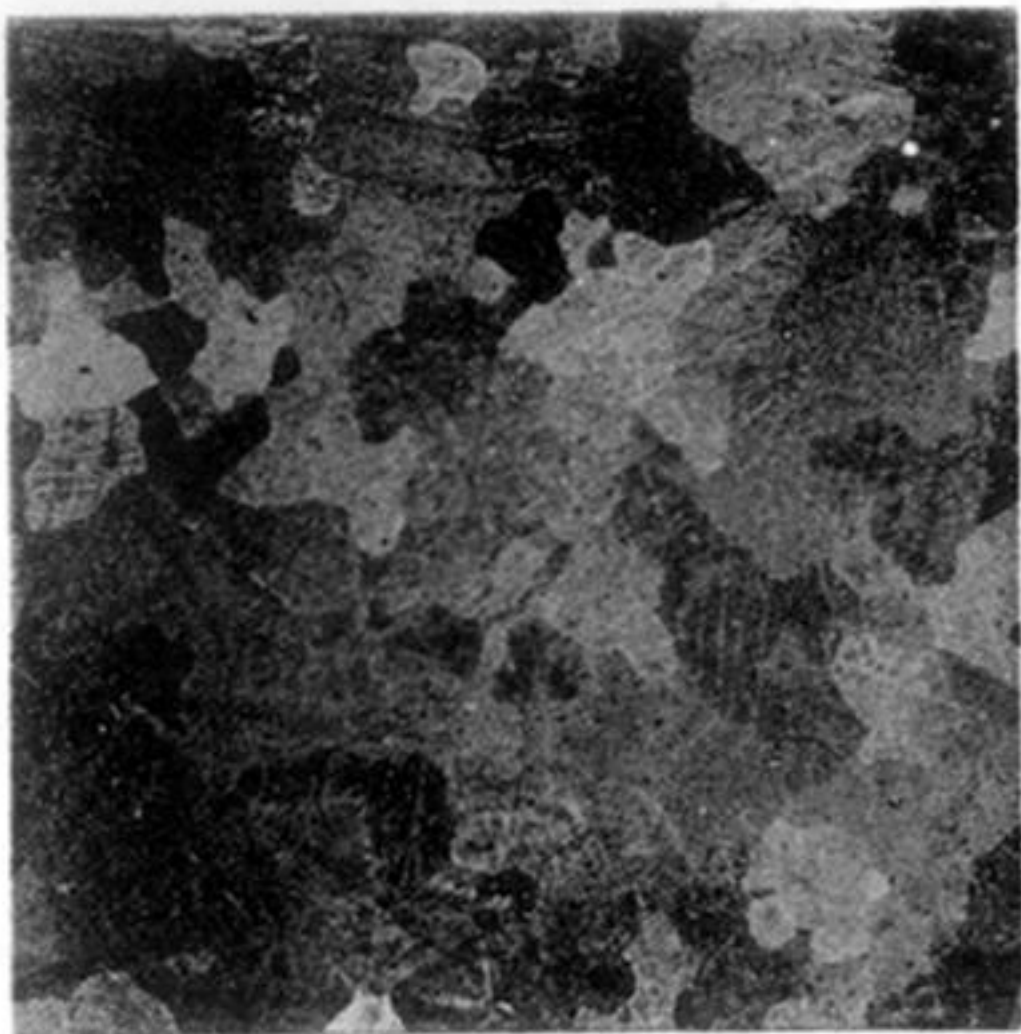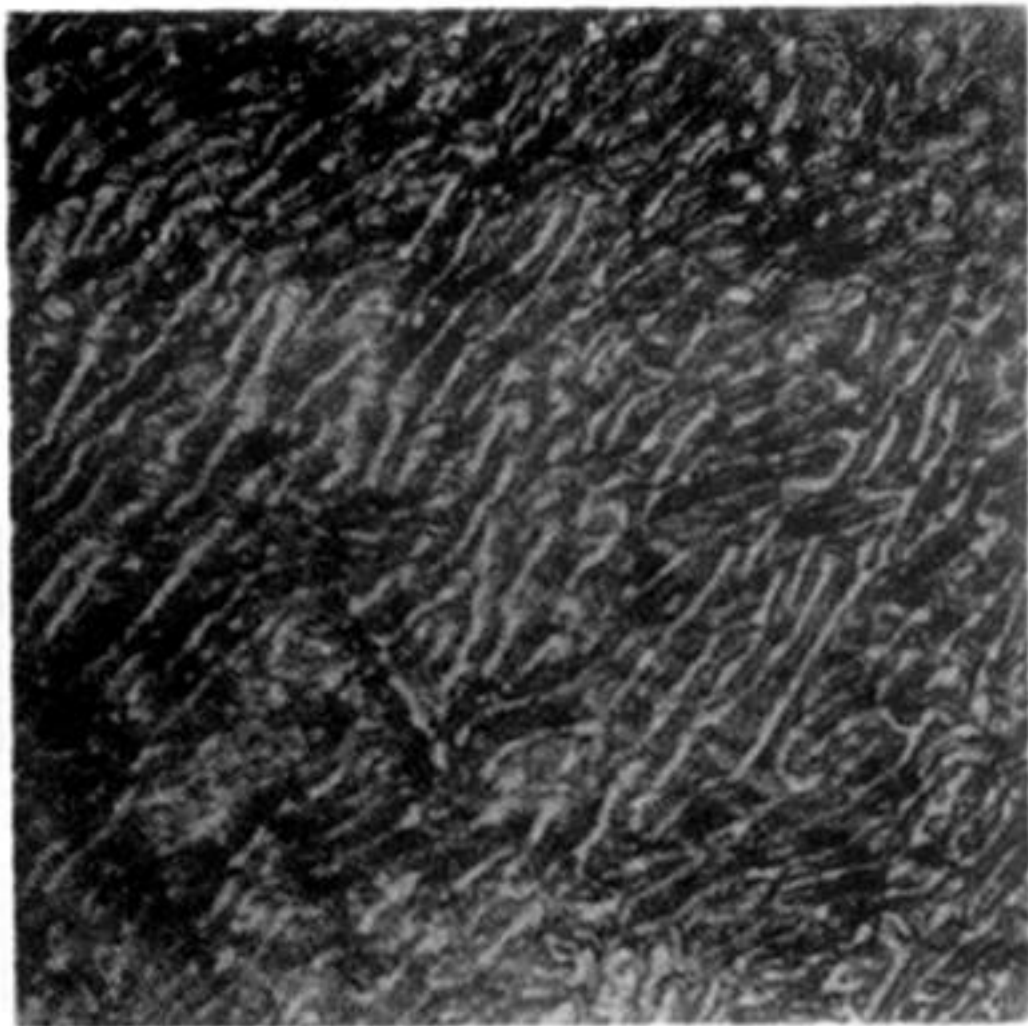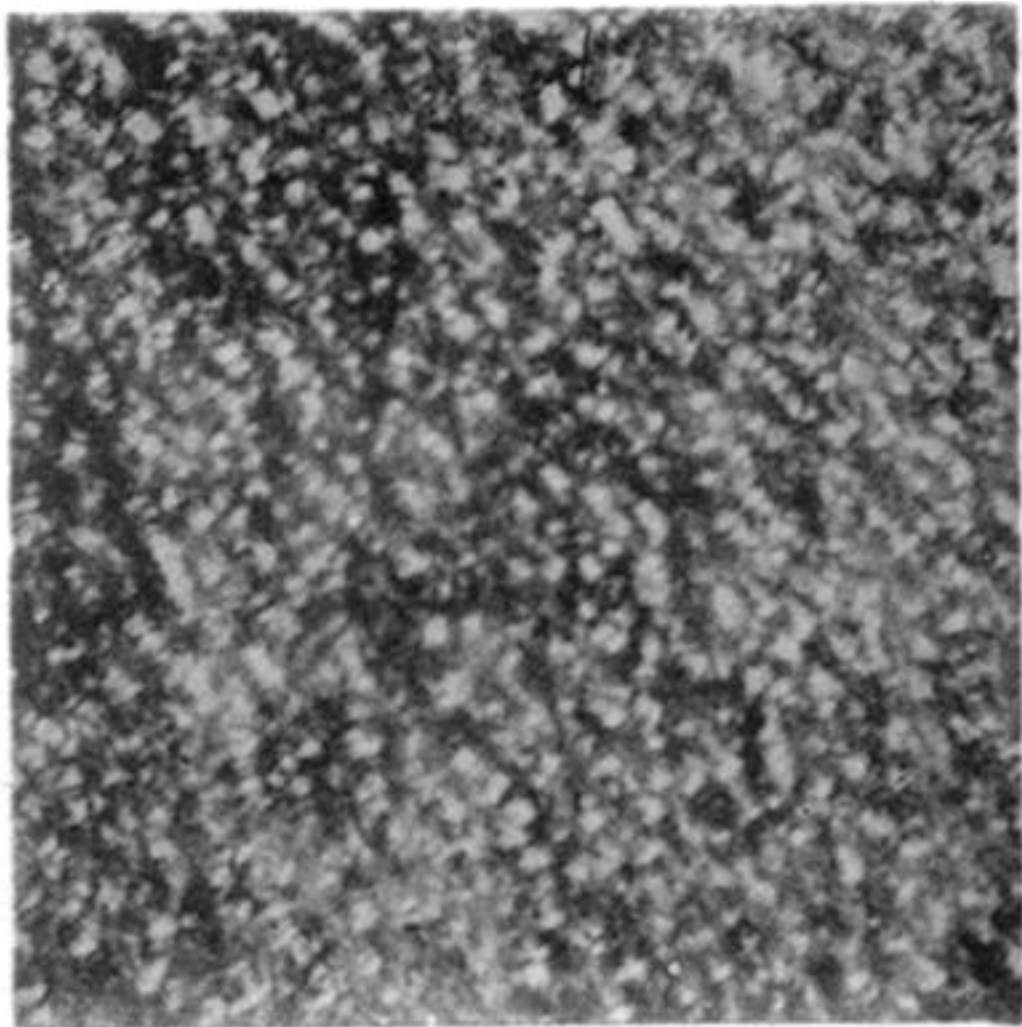

Fig. 5.   27 per cent Gold, 73 per
cent Copper. x 4.5D

Fig. 6.   Eutectic portion of 27 per cent
Gold, 73 per cent Copper. x 6320D

FIG. 2.

Fig..1.   Surface of an Ingot of Standard
Gold, (unetched) x 4. 5D.

Fig. 2.　Standard Gold.　x 4.5D

Fig. 3. Eutectic, (80 per cent Gold, 20 per cent Copper) x 1580D.

Fig. 4.   Standard Gold.   x 1580D.

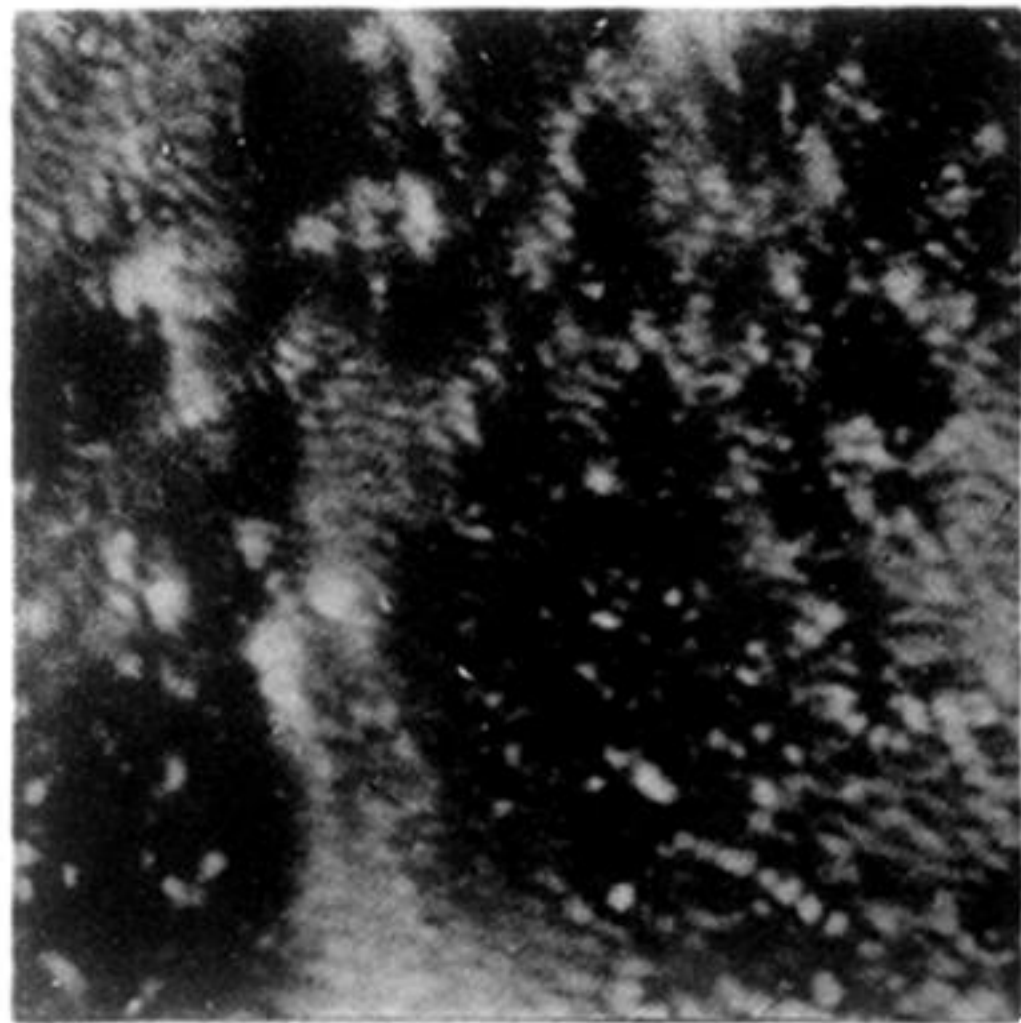Fig. 5. 27 per cent Gold, 73 per
cent Copper. x 4. 5D

Fig. 6.   Eutectic portion of 27 per cent
Gold, 73 per cent Copper. x 6320D